EFFECT OF MOISTURE AND ADDITIVE ON RADON EXHALATION RATE FROM CONCRETE

AMIT KUMAR1,2, R.P. CHAUHAN1

1Department of Physics, National Institute of Technology, Kurukshetra-136119, India
2Department of Applied Sciences, Asia Pacific Institute of Information Technology, Panipat, India
E-mail: amit.vera@gmail.com, chauhanrpc@gmail.com

Received October 3, 2014

Building materials are significant contributor of indoor radon after the soil. The indoor radon level depends upon the radon flux coming out from wall, ceiling and roofs of the dwellings. The radon flux from the wall that in most cases is concrete depends upon many factors like radium content, porosity, moisture, composition of mix design for construction of concrete and age of concrete. The present work is a study of effect of various factors on the radon exhalation rate from concrete. The measurement of radon exhalation rates from the concrete was determined by active technique using continuous radon monitor. The results show that the radon exhalation rate decreased with decrease of porosity of concrete and increase of moisture content after a particular value. The various additives like silica fume and fly ash may cause the decrease of radon exhalation up to optimum value, after that it increases. The radon exhalation rate also varied with age of concrete.

Key words: Radon exhalation rate; Moisture content; Modified concrete; Porosity.

1. INTRODUCTION

Indoorn radon gas is one of the most significant contributors of the inhalation dose received by the human from background radiation. The long half life of radon (3.824 days) makes it important to study in residential building and work places. The heaviour nature of radon causes its higher accumulation in poorly ventilated dwellings [1]. The building materials are the second most important source of indoor radon in dwellings after soil.

The radium present in the grain of building materials emanate radon which comes out to indoor environment by diffusion and advection. The fraction of emanated radon from gain to pore and pore to environment depends upon many factors like porosity, moisture, age of concrete, radium content, composition of mix design for construction of concrete. The effect of radon exhalation rate from building materials on indoor radon concentration was discussed in Kumar et al. [2]

The estimation of indoor radon concentration was made using various models and validated by the experimental measurements. The effect of the grain size, radium content and free volume over the sample in closed accumulator technique using AB-5 (Pylon) was studied by Chau et al. [3]. The ratio of container volume to sample volume has significant influence on the measured radon exhalation rate. The radon exhalation rate from the building materials of decorative use was measured by Saad et al. [4] and found that non-dried porous materials have low radon exhalation than the dried materials in case of structural building materials.

Kovler et al. [5] performed the measurement of radon exhalation rate from the concrete containing fly ash and reported that the radon exhalation rate from concrete depends upon the water binder ratio and radium content of the fly ash. Shweikani and Raja [6] reported that the radon exhalation rate from building materials varied with their type and origin. Kovler [7] studied the hardening of concrete and radon exhalation rate from the construction site and predict the radon concentration under different ventilation rate of chamber. The leakage/ventilation rate of chamber play important role for determining the radon exhalation rates [8, 9]. The radon exhalation rate of the samples depends upon the radium content of the samples studied by Shoeib and Thabayneh [10].

The radon exhalation rate from building materials also depends on radon diffusion length and thickness of the concrete/wall [2, 11–13]. The other factors that affect the radon exhalation rate are moisture content, additives, porosity and age of concrete. The present study considers the dependence of radon exhalation rate on these factors. For study purpose the concrete modified with different additive and cement ratio were prepared in laboratory. The measurement of radon exhalation rate was performed by active radon monitor (SRM).

2. MATERIALS AND METHODS

2.1. PREPARATION OF CONCRETE TILES

The concrete tiles of dimension 21.5 cm × 21.5 cm × 2.3 cm were prepared with mix design as per ASTM standards [14]. A part of cement is replaced with silica fume and fly ash as additives. The details of the procedure for preparation of concrete and mix design were described elsewhere [15]. The porosity of concrete tiles was determined by water absorption test [16]. To study the effect of moisture the concrete tiles were dried in the oven for different time and moisture content was calculated by difference in the mass of concrete with that of totally dried concrete.

2.2. MEASUREMENT OF RADIIUM CONTENT

For the measurement of radium content, the samples were crushed in a vibrating ball machine to convert the samples in small grain size and dried in an
Effect of moisture and additive on radon exhalation rate from concrete

oven at 110° C for 48 hours to remove the moisture. The samples were then sealed in the containers having dimension 6.2 cm height and 5.8 cm diameter for 28 days so as to attain the secular equilibrium between the $^{226}$Ra, $^{232}$Th and their progenies. The measurement of radium, thorium and potassium contents of the sample were carried out on HPGe gamma radiation detection unit. The detector efficiency for particular gamma peak was determined by the IAEA standard sources (RGU, RGTh and RGK). The humidity and temperature of the laboratory were kept constant during the counting for standard source as well as samples under study. Since the activity of the samples was low, thus, the counting was done for long period of 2,50,000 sec. For the estimation of radium energy peaks 186.1 keV from $^{226}$Ra, 609.4 and 1764.5 keV from $^{214}$Bi were used (17).

2.3. MEASUREMENT OF RADON EXHALATION RATE

The measurements of radon exhalation rates from samples were carried out by hermetically enclosing the sample with in accumulator [18–19]. The accumulator was a stainless steel cylinder with an inner height of 50 mm, and a diameter of 300 mm with provision to attach a Lucas cell coupled photomultiplier tube at upper side as shown in Figure 1.

Fig. 1 – Experimental set up used for radon exhalation study.

The growth of radon in container was measured by scintillation radon monitor (SRM). The detail of the measurement procedure of radon exhalation rate is described in Kumar et al. [2]. A concrete tiles under study were placed in the exhalation chamber and the growth of radon in the chamber was determined by the scintillation radon monitor. The radon concentration inside the exhalation chamber
increases with the time and reaches a saturation value. The radon growth data then fitted in following equation:

\[
C = \frac{E A}{V \lambda_e} (1 - e^{-\lambda_e t}) + C_i e^{-\lambda_e t},
\]

where \( E \) represents the radon surface exhalation rates in (Bq/m²/h), \( A \) and \( V \) represent the surface area of sample and effective volume of the chamber including the volume of the scintillation cell. \( \lambda_e \) represents the effective decay constant, which is sum of radon decay constant and chamber leakage rates if any, \( C_i \) is initial radon concentration in chamber.

3. RESULTS AND DISCUSSION

3.1. RADIUM CONTENT FROM SAND, CEMENT, FLY ASH AND SILICA FUME

The radium content of raw building materials (sand, cement, fly ash and silica fume) was determined by HPGe gamma detector is shown in Figure 2.

![Figure 2 - Radium content from various raw building materials.](image)

The radium content of fly ash is higher than that of sand, silica fume and cement. This may be because the fly ash is the waste product of coal thermal power plant in which coal is used as fuel. Upon burning of coal in the thermal power
plant, the radium content in fly ash is concentrated. Thus the radium content in the fly ash is expected to be higher compared to other building materials. When this fly ash is added to cement it may cause an increase in radium content in mix design and hence an enhancement of radon exhalation rate.

3.2. RADON EXHALATION RATE FROM MODIFIED CONCRETE

The radon exhalation rate from concrete at the age of 28 days (reported in our earlier publication [16] for silica fume is slightly higher than that at age of 60 days for below 20% replacement). This increase in radon exhalation rate was observed for the 20% replacement of silica fume up to which the formation calcium silicate hydride bonding is pronounced during concrete formation. With increase in the age of concrete the CSH bonding increases causing increase in porosity and decrease in radon exhalation rates. The variation of radon exhalation rate with addition of fly ash and silica fume at the age of 60 days is shown in Figure 3.

![Figure 3](image)

**Fig. 3 – Variation of radon exhalation rate with substitution of fly ash and silica fume.**

The radon exhalation rate of concrete substituted with silica fume continuously decreased with increase of its content. This may be attributed to fact that the increase in silica fume content in cement cause increase in silica content. This may causes increase in the calcium silicate hydrate linkage (CSH) and hence decrease in porosity [16]. Due to decrease in porosity, the volumes of the pore space through which radon atom can diffused out of the concrete decreases and hence the radon exhalation rate is decreases. On the other hand the radon
exhalation rate from the concrete substituted with fly ash, first decrease with increase in fly ash content and then increase. With increase of fly ash content the porosity of concrete decreases and radium content increases because the radium content of fly ash is higher than cement discussed in section 3.1 and shown in Figure 2. Initially the effect of radium content is small as compare to porosity thus a decrease in exhalation is observed up to 30 %, beyond which increase in radon exhalation is attributed as increase in radium content. The porosity of the concrete depends upon the quantum of CSH linkage, higher the CSH linkage, smaller the porosity. With increase in the fly ash in cement caused decrease in the porosity by formation of additional CSH linkage thus, reduced the radon surface exhalation rate. This means that the porosity dominates for radon surface exhalation up to 30 % of fly ash addition. Beyond 30 % fly ash substitution the increase in radium content dominate over the decrease in porosity and hence caused the increase in the radon surface exhalation rates.

3.3. EFFECT OF MOISTURE AND RADIUM CONTENT ON RADON EXHALATION RATE

The trend of radon exhalation mentioned in section 3.2 is valid for completely dried concrete. However the moisture present in the concrete play important role in measurement of radon exhalation rates. The effect of moisture on radon exhalation rates of concrete was measured by drying the wet concrete for different time period so that the concrete had different moisture contents. The percentage moisture contents of concrete were determined by the difference in the mass of wet and dry concrete. The variation of radon exhalation rates with moisture content is shown in Figs. 4 and 5 for fly ash and silica fume substituted concrete.

![Fig. 4 – Variation of radon surface exhalation rate with moisture content from fly ash modified concrete.](image-url)
The radon exhalation rates of fly ash concrete first increased with increase of water content up to 17% after that it decreases. On the other hand, for silica fume concrete, the increase is restricted up to 9% before increase. This shows the dependence of radon emanation and exhalation from a grain and concrete blocks and tiles upon the moisture content. The radon emanates from its parent nuclides through alpha decay of radium-226. The recoils of alpha particle push the radon in opposite direction with a recoil length 30–50 nm in solid grain [20–22]. For the recoiled radon atom there are following possibilities (i) the recoiled radon atom may be retained inside the same grain due to small recoil length (ii) it may enter into the neighbouring grain (iii) it may emanate out from grain and enter in pore space. If radon stays in the pore space then it can be transferred to air by diffusion or advection. The recoil length of radon in water is smaller than that of air, thus due to presence of water in pore space more number of atoms trap in water present in pores rather than to enter in another grain. This may cause increase in the radon exhalation rate of materials. But with increase in moisture contents, the water in pore space of material increases, causing more radon atoms to decay in water rather than escaping out of the material and hence shows a decrease in exhalation rates. The most important factor that affects the radon exhalation rate is radium content of building materials. The radium content of sample was estimated by knowing the radium content of sand, cement fly ash and silica fume using HPGe gamma ray spectrometry. The variation of radon exhalation rate from radium content of concrete is shown in Figure 6.
4. CONCLUSIONS

The building materials are important contributor of indoor radon after soil and depend upon many factors. The radon exhalation rate of concrete substituted with fly ash and silica fume have different radon exhalation rate depending upon the limit of their replacement with cement. For concrete with silica fume and fly ash the radon exhalation rate first decreases and then increases in case of fly ash. The higher radium content in fly ash compared to cement and sand should be taken into consideration when use for substitution of cement. The increase in the moisture content causes decrease in the radon exhalation up to 9 and 17% for silica fume and fly ash. After that it decreases. Thus care should be taken about the moisture content of sample when calculating specific radon exhalation rate. The increase in radium content of the sample also causes increase in radon exhalation rate. The use of building materials with higher radium content should be avoided for construction purpose.

Acknowledgements. The authors are thankful to Board of Research in Nuclear Science, Department of Atomic Energy, Mumbai, India, for providing the instrument for carrying out this work. The help received from Civil Engineering Department, National Institute of Technology, Kurukshetra, India is also thankfully acknowledged.

REFERENCES

15. Chauhan, R.P., Kumar, A. Atmospheric environment, 81, 413-420 (2013).